

## ACCUMULATION OF HEAVY METALS IN SOIL AND VEGETABLES DUE TO WASTEWATER IRRIGATION IN A SEMIARID REGION OF RAJASTHAN, INDIA

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### ABSTRACT

Irrigation with wastewater has become a common practice in the arid and semi arid regions due to scarcity of fresh water. This leads to adverse effect on soil and human health. The present study was conducted to evaluate the effect of wastewater irrigation on agricultural fields at two wastewater irrigated sites at Jaipur city in Rajasthan, India. The aim of the study was to estimate the accumulation of heavy metals (Pb, Cd, Cr and Ni) in soils and vegetables due to wastewater irrigation. The study revealed that concentration of Pb and Ni in soil was under permissible limits whereas the concentration of Cr and Cd was found to be above the permissible limits given by WHO. Vegetables (*Spinacia oleracea* (spinach), *Solanum lycopersicum* (tomato), *Abelmoschus esculentus* (lady finger) and *Solanum melogena* (brinjal) were found to accumulate all the heavy metals in high concentration. Among all the vegetables, spinach was found to accumulate highest concentration of all the metals. This study suggests that the consumption of vegetables grown on wastewater irrigated site can prove to be hazardous for human health leading to several fatal diseases including cancer.

**KEYWORDS:** Contamination, Heavy Metals, Vegetables, Wastewater Irrigation

### INTRODUCTION

The use of wastewater in agricultural fields is a common practice worldwide and especially in the arid and semiarid regions where there is shortage of fresh water resources for production of crops to cater the rapid growing population. Farmers are using the wastewater for irrigation for two reasons; firstly due to shortage of fresh water and secondly due to the presence of some nutrients in the wastewater which is known to improve the production of crops. Unaware of the fact that the wastewater also contains toxic heavy metals it is indiscriminately used in the agricultural fields. Several studies have shown that wastewater irrigation significantly contributes to the heavy metal content of soil (Mapanda *et al.*, 2005; Nan *et al.*, 2002). This accumulation of heavy metals is known to degrade the quality of soil and pose deleterious health effects when transferred in the food chain (Rattan *et al.*, 2002). Use of sewerage water and industrial effluents is known to be the main reason for accumulation of heavy metals in vegetables along with ariel deposition from thermal power plants and abundant use of pesticides in the agricultural fields (Gupta *et al.*, 2008).

Heavy metals are the most important type of contaminant that can be found on the surface and in the tissues of the crops grown in the wastewater irrigated agricultural fields. Certain metals are required for the growth of plants in very low concentration but at higher concentrations they are known to cause soil degradation and toxic effects on human health when consumed with food. Heavy metals are of major concern due to their long retention time. lead (Pb) is found to have a soil-retention time of 150–5,000 years and is reported to maintain a high concentration for as long as 150 years after

entering the soil (Nanda kumar *et al.*, 1995) The half-life of lead varies from about a month in blood, 1-1.5 months in soft tissue, and about 25-30 years in bone (ATSDR 2007). Young children, especially those under the age of 6, are at greatest risk for lead poisoning. A child's developing brain and central nervous system are more susceptible to the toxic effects of lead since the blood-brain barrier is not complete until approximately 36 months of age and during that time there is extensive neuron development. Many of these effects are irreversible. The biological half-life of cadmium (Cd) is about 18 years. Cd is known to cause damage to the kidney in the form of cadmium – metallothionein(Cd-MT) (Barbier *et al.*, 2005), adversely affects the productions of progesterone and testosterone (Piasek and Laskey 1999) as well as the IARC (International Agency for Research on Cancer) has classified cadmium as a human carcinogen. Similarly, the Department of Health and Human Services has determined that metallic nickel may reasonably be anticipated to be a human carcinogen and nickel compounds are known to be human carcinogens. Similarly, IARC classified metallic nickel in group 2B (possibly carcinogenic to humans) and nickel compounds in group 1 (carcinogenic to humans). Ingestion of Cr(VI) produces gastrointestinal burns and hemorrhage, liver damage, and kidney damage that may lead to death. Reduced sperm count is also observed due to Cr (VI) intake (ATSDR, 2000).

Heavy metals not only affect human health but are also known to affect soil. They show detrimental effects on decomposition and nutrient mineralization as they destruct the microbial flora of soil, leading to loss of fertility (Berg *et al.*, 1991). Thus it is evident that contamination of soil due to heavy metals not only affects human health but also adversely affect soil health leading to depletion of this important natural resource. The present study was thus conducted keeping in view the extensive use of wastewater in agricultural practices and its adverse effects on human health.

## MATERIALS AND METHODS

### Study Area

The study area of the present work, Jaipur, a semi arid region is the capital city of Rajasthan state situated at (26.920N 75.820 E) with an altitude of 431 m above the sea level occupying an area of 11,117.8 Km<sup>2</sup> situated in the eastern border of Thar desert. The city is well known for its *Sanganeri* prints (traditional printed fabric). Clusters of printing units are established in the sanganer area of the city, one of our study site. Chemical dyes are used in these printing units and disposed off without treatment in the common drainage, *Amanisha nala*. The *Amanisha Nala* collects wastewater from several other industrial sources as well including the municipal sewage without treatment. Vegetables are grown all along the *nala* using the untreated wastewater as a source of irrigation. Common vegetables grown in these areas are Spinach, Tomato, lady finger and brinjal.

### Sampling and Analysis

For the purpose of assessing the adverse effect of wastewater irrigation on soil and accumulation of metals in vegetable, two wastewater irrigated sites i.e agricultural fields at Sanganer and Durgapura and a control site where bore well water was used for irrigation were selected . The sampling and analysis was conducted between January to June 2014. Samples of wastewater from the *Amahnisha Nala*, soil samples(0-15 cm depth ) and four vegetables Spinach, Tomato, lady finger and brinjal were collected in triplicates in order to estimate the concentration of metals(Cd, Ni, Pb and Cr). Wastewater samples collected from *Amanisha nala* were pretreated with concentrated HNO<sub>3</sub> to prevent microbial degradation of heavy metals. For analysis, 50 mL of wastewater samples were digested with 10 mL concentrated HNO<sub>3</sub> at 80°C (APHA 1985). Soil samples were air-dried, crushed and passed through 2-mm mesh sieve and stored at ambient temperature prior to analysis. The freshly harvested mature vegetables were brought to the laboratory and washed primarily

with running tap water, followed by three consecutive washings with distilled water to remove the soil particles. Samples were cut into small pieces and dried in oven at 80°C for 48 h and then ground to powder. 0.5 g each of soil and vegetable samples were digested (wet acid digestion) with concentrated HNO<sub>3</sub>, H<sub>2</sub>SO<sub>4</sub> and HClO<sub>4</sub> (5:1:1) at 80°C (Allen *et al.* 1986) until the solution became transparent. The digested samples of water, soil and vegetables were filtered through the Whatman No.42 filter paper and the filtrates were diluted to 50 mL with distilled water. All reagents used were Merck, analytical grade (AR) including Standard Stock Solutions of known concentrations of different heavy metals.

Heavy metal concentrations of wastewater, soil and vegetable samples were estimated by Atomic Absorption Spectrometer (Thermo). Blank samples were analyzed after seven samples. Concentrations were calculated on a dry weight basis. All analyses were replicated three times. The accuracy and precision of metal analysis were checked against NIST-SRM, 1570 for every heavy metal. The results were found within ±2% of certified values. All the data are expressed as mean± standard deviation. The data was analyzed using one way analysis of variance (ANOVA) followed by LSD post hoc test. All the analysis was performed using SPSS software (Version 20)

## RESULTS AND DISCUSSIONS

### Heavy Metals in Wastewater

The mean concentrations obtained(Table1) from the analysis of wastewater samples from *amahnisha nala* flowing near study sites and the bore well from the control site show that all the metals in the wastewater are higher than the safe limit given by WHO whereas metals were not detected in bore well water samples. The mean concentration of metals were found to be higher at Sanganer as compared to Durgapura. Cr showed the highest concentration(7.14 mg/l) at Sanganer whereas Ni was found in lowest concentration (0.89 mg/l) at durgapura. The reason for higher metal concentration at Sanganer can be the disposal of untreated wastewater from the cluster of printing units in this area. Whereas at Durgapura, addition of wastewater from other sources (municipal wastewater) dilute the concentration of metals in water. Cr showed the highest concentration at both the sites followed by Pb, Cd and Ni. Significant difference (*p*< 0.05) was observed for Cd at Durgapura whereas Pb, Cr, and Ni showed significant difference at (*p*<0.01) level at both the sites. The mean concentration of Cr, Cd, Ni and Pb was found several folds higher in the present study as compared with the results of wastewater analysis reported by Sharma *et al.*, 2007 at Varanasi where the mean concentration of metals (mg/l) were Cr (0.053), Cd (0.016), Ni(0.012).Singh *et al.*, 2012 reported higher levels of Pb(7.5mg/l) and Ni(401 mg/l) and lower level of Cd(2.1 mg/l) in wastewater used for irrigation at Nagpur as compared to the results of present study. The reason for this can be the quality of wastewater discharged from the industries. In the present study maximum quantity of wastewater is discharged from the textile printing units using chemical dyes, which are a source of heavy metals.

**Table 1: Mean Concentration of Heavy Metals (mg/l) in wastewater  
(Collected from Amahnisha Nala) used for Irrigation (n=48)**

	Cr (Mg/L)	Cd(Mg/L)	Ni(Mg/L)	Pb(Mg/L)
<b>Durgapura</b>				
Mean± SD	5.0**±1.08	1.95*±0.25	0.89**±0.73	2.08**±0.39
<b>Sanganer</b>				
Mean± SD	7.14**±0.54	3.43**±0.87	2.18**±0.97	3.6**±0.90
<b>Control(Bore Well Water)</b>				
	ND	ND	ND	ND
Safe Limits <sup>a</sup>	0.1	0.01	0.2	0.05

**Source:** Pescot (1992), n= Number of Samples, \*Significant at 0.05 Level, \*\*Significant at 0.01 Level

### **Heavy Metals in Wastewater Irrigated Soil**

Analysis of soil indicates that continuous application of untreated wastewater in the study areas has led to accumulation of heavy metals in soil as compared to the bore well water irrigated site where heavy metals (Cr, Cd, Ni, Pb) were below detectable level(Table 2). The concentration of Ni and Pb were found under the permissible limits but were high enough to accumulate in the vegetables whereas Cd and Cr were found in higher concentration as compared to the permissible limits. All the heavy metals showed a significant difference at  $p<0.01$  level. The concentration of all the metals was found higher at Sanganer as compared to Durgapura. The mean concentration of Cd (14.7mg/kg) and Pb(33.86 mg/kg) recorded in the present study at Sanganer and Cd(13.62mg/kg) and Pb(31.34 mg/kg)at Durgapura are lower than the concentration Cd(24.66mg/kg) and Pb (52.72mg/kg) reported at Dehradun reported by Rai *et al.*,(2012). Whereas the mean concentration of Cd(14.7 mg/kg), Cr(38.39) and Ni(30.08mg/kg) observed in the present study at Sanganer are higher than the reported concentration of Cd(3.12),Pb(21.95),Ni(23.65)in wastewater irrigated site at Varanasi(Singh *et al.*, 2010).Whereas many fold higher concentration of Pb (441.7 mg/kg) and Ni (276.6 mg/kg) were reported by Qishlaqi *et al.* (2008) at Iran as compared to the concentrations obtained in the present study at wastewater irrigated sites.

**Table 2: Mean Concentration of Heavy Metals in Soil N=36**

	Cr	Cd	Ni	Pb
<b>Durgapura</b>				
Mean( $\pm$ ) SD	33.53**( $\pm$ )1.31	13.62**( $\pm$ )1.82	27.95**( $\pm$ )1.62	31.34**( $\pm$ )1.81
<b>Sanganer</b>				
Mean( $\pm$ ) SD	38.39**( $\pm$ )2.50	14.7**( $\pm$ )1.01	30.08**( $\pm$ )1.47	33.86**( $\pm$ )2.38
<b>Control</b>				
	ND	ND	ND	ND
<b>Safe Limits <sup>a</sup></b>	--	3-6	75-150	250-500

<sup>a</sup>Awashthi 2000, \*\*Significant at 0.01 Level, ND- Not Detected

### **Heavy Metal Concentration in Vegetables**

Accumulation of metals in crops grown on contaminated soils has been found in several research studies worldwide. The accumulation of metals in vegetables depends on several factors like concentration of metals in soils, type of vegetable (leafy, tuber, fruit), soil organic carbon % and pH of soil. pH playing a significant role where low pH enhance the bioavailability of metals. The concentrations of heavy metals in the vegetables in the study area of the present research work were found to be much above the permissible limits given by WHO/FAO and showed a significant difference at  $p<0.01$  level. Heavy metal concentrations varied among different vegetables sampled which may be attributed to different absorption capacity of vegetables for different heavy metals (Zurera *et al.* 1989). Vegetable samples from Sanganer showed the maximum concentration of all metals as compared to the samples of Durgapura area. This can be attributed to the higher concentrations of all metals in wastewater and soil at Sanganer area. Among the four vegetables sampled, Spinach a leafy vegetable was found to accumulate maximum amount of all the heavy metals.

### **Chromium**

The Cr concentration in the present study was found to be highest (17.26 mg/Kg) in spinach followed by tomato(15.26 mg/kg),brinjal (5.1mg/kg) and lady finger (4.25 mg/Kg) at wastewater irrigated fields of Sanagner site. These levels are much higher than the levels (0.1-0.2 mg/Kg) permissible by WHO. Higher levels of Cr in spinach as compared to the present study were reported by several other studies at wastewater irrigated sites like (13.91 mg/kg)

observed by Prabu (2009) at Ethoipia and (11.21 mg/Kg) at Agra by Kumar (2013). Many fold higher levels of Cr (96.30 mg/kg) were reported by Gupta *et al.*, (2008) in spinach at Titagarh in West Bengal and (70.79mg/Kg) in spinach reported by Ramesh and Murthy (2012) at Bangalore

Mukherjee and Mishra (2008) reported slightly lower value (9.28 mg/Kg) of Cr as compared to the present study in spinach during summer whereas in winter, values (6.06 mg/kg) were reported. Lower Cr values (0.194 mg/Kg) for Spinach were reported by Lawal and Audu(2011) at Nigeria during dry season and further lower level (0.63 mg/Kg) in rainy season. The Cr concentration in tomato in present study (15.26mg/kg) at Sanganer and (12.64 mg/kg) at Duragpura are higher than the values reported by several other studies. Cr in tomato(4.91 mg/Kg) was reported by Prabu (2009), (0.35 mg/kg) Lawal and Audu (2011), (0.38mg/kg) Liu *et al.*, (2006) and (0.2 mg/Kg) Beenerji *et al.*, (2011). The Cr level (5.1 mg/Kg) in Brinjal and(4.25 mg/Kg) in lady finger in the present study is lower than reported by Lawal and Audu(2011) (6.66 mg/Kg) and a level(5.45 mg/Kg) respectively at Nigeria. Whereas lui *et al.*, ( 2006) reported a very low concentration of Cr (0.35 mg/Kg) in brinjal as compared to the present study.

### Cadmium

The concentration of Cd in all the vegetables were found to be above the permissible limits (0.1-0.5 mg/kg) given by WHO at both the sites studied with higher concentration at Sanganer. The levels of Cd for different vegetables Spinach (4.56 mg/kg), Tomato (3.29 mg/kg), Lady finger (2.62 mg/kg) and brinjal (4.47 mg/kg) found in the present study are comparable with the results reported by Mukherjee and Mishra (2008) in Varanasi city with a value of (5.20 mg/kg) in spinach. Qishlaqi *et al.*, (2008) also observed accumulation of Cd (3.59 mg/Kg) in Spinach at wastewater irrigated site and very low level of metals in tube well irrigated sites at Iran. Similar results were observed in vegetables by Bahemuke and mubofu (1999); Nambiar and Sinha (2006) and Alloway (1999). Very low levels of Cd (0.12 mg/Kg) for Spinach, (0.09 mg/Kg) for Tomato and (0.11 mg/Kg) for Ladyfinger are reported by Rapheal *et al.*, (2011) at Nigeria as compared to the values obtained in the present study. Similarly low levels of Cd in tomato(0.085 mg/Kg) and Brinjal (0.11 mg/Kg) were observed by Liu *et.al* (2006) in China in polluted water irrigated sites. At Ethopia the levels of Cd for Tomato(0.115mg/kg) and (0.285 mg/Kg) were observed by Prabu *et al.*,(2009) and (0.9mg/kg) at Agra was observed by Kumar (2013). In Bangalore the value of Cd for Spinach was found to be (0.81 mg/Kg) by Ramesh and Murthy (2012) in the wastewater irrigated site and Cd was below detectable level in all the reported studies in ground water irrigated site conforming the results of the present study. Lower levels of Cd were also observed in all the vegetables by Parashar and Prasad 2013 at Agra Cd concentration (1.09mg/kg) in spinach, (0.61 mg/kg) in tomato, (0.68 mg/kg) in lady finger and (0.47 mg/kg) in brinjal. In contrast to the present study higher level of Cd in Spinach was observed to be (14.58 mg/Kg) at Titagarh in West Bengal reported by Gupta *et al.*, 2008.

### Lead

All the vegetable samples analysed, showed very high concentration of Pb at all the wastewater irrigated sites which is much above the permissible limits (0.1 mg/kg) in vegetables given by the WHO. The maximum concentration (15.51 mg/kg) of Pb was observed in spinach followed by tomato (11.39 mg/kg),(3.08 mg/Kg) in brinjal and(5.21 mg/Kg) in lady finger at Sanagner. A higher concentration(9.35 mg/Kg) of Pb was reported by Lone *et al.*, 2003 in Lady finger and a much higher value(97.2 mg/kg) at Lahore observed by Khan *et al.*, 2013 in Spinach and (39.39 mg/kg ) in tomato from sewage irrigated fields as compared to the present study. Parashar and Prasad 2013also reported higher levels of Pb at Agra (31.42 mg/kg) in spinach,(24.51mg/kg) in tomato,(23.34 mg/kg) in Lady finger and (27.72 mg/kg) in Brinjal. Siddique *et*

al. 2014 reported a much higher level of Pb in vegetables at Faislabad. They found (99.8 mg/kg) Pb in leafy vegetables and (83.7 mg/kg) Pb in fruit vegetables which are much higher than the values observed in the present study. In contrast to the observations of present study, lower levels of Pb were observed by Rapheal *et al.*, 2011 in Nigeria where Pb concentration in Spinach was (0.22 mg/Kg), in Tomato(0.23mg/Kg) and Lady finger(0.12 mg/Kg) the reason can be lower concentration of Pb in soil and higher values of soil pH and OC % reducing the phytoavailability of the metal.

### Nickel

All the vegetables grown on wastewater irrigated site showed a many fold higher value of Ni in the present study as compared to the permissible limit(1.5 mg/kg) given by WHO. Maximum concentration (5.77 mg/kg) was found in Spinach followed by the value(5.38 mg/Kg) in Lady finger, (5.78 mg/kg) in brinjal and (3.47) in Tomato grown at Sanganer site. In contrast to the result obtained in the present study, Singh *et al.*, 2008 reported much higher values (39.25 mg/Kg) at Varanasi, lone *et al.*, (2006) observed (28.00mg/kg) of Ni in spinach and (20.80 mg/kg) in lady finger from the wastewater irrigated site as well as Sharma *et al.*, 2006 reported a maximum value(15.5 mg/kg) at Dinapur. Whereas very low values of Ni (3.0 mg/Kg) were reported in wastewater irrigated site in Spinach at Hyderabad by Chary *et al.*, (2008).

The variation in the concentration of metals in vegetable grown in wastewater irrigated sites is dependent on various factors like concentration of metal in soil, concentration of metals in soil solution, pH and organic carbon in soil. Thus even if concentration of metals in soil is high, plant uptake may be very low if metal concentration in soil -solution is less. Low pH enhances the availability of metals in soil solution. Another factor, the organic carbon % in soil helps to bind the metals on soil particles making less metal available in soil-solution and in turn less uptake by plants. The variations in absorption of metals in plants through roots and their further translocation within the plant parts, edible parts of vegetables is also dependent on the plant species (Vousta *et al.* 1996).

**Table 3: Mean Concentration of Heavy Metals in Vegetables**

Study Sites	Cr(mg/kg)	Cd(mg/kg)	Ni(mg/kg)	Pb(mg/kg)
<b>Tomato</b>				
Durgapura	12.64**±3.16	1.72**±0.63	1.77±1.28	8.54**±0.05
Sanganer	15.26**±1.76	3.29**±1.45	3.47**±1.26	11.39**±1.15
<b>Spinach</b>				
Durgapura	15.67**±1.56	2.25**±0.72	5.21**±1.03	13.36**±1.15
Sanganer	17.26**±2.31	4.16**±2.17	5.77**±1.38	15.51**±1.05
<b>lady Finger</b>				
Durgapura	2.68±0.41	1.73±0.99	4.21**±0.85	2.13**±0.28
Sanganer	4.25±1.37	2.62±1.19	5.38**±1.28	3.08**±0.90
<b>Brinjal</b>				
Durgapura	2.92**±1.11	2.44**±0.83	4.42**±1.09	3.74**±1.02
Sanganer	5.1**±0.62	4.47**±1.23	5.78**±0.91	5.21**±0.53
Control site	BDL	BDL	BDL	BDL
Safe limits <sup>b</sup>	0.1-0.2	0.1-0.5	1.50	0.1-0.3

<sup>b</sup>Limits given by WHO/FAO (Codex Standard 193-1995)

## CONCLUSIONS

The higher metal concentration in vegetables found in the present study is a serious concern as these metals are known to cause irreversible damage to the human body by disrupting of various systems of the body like Cd adversely affects the kidney and liver, Cr affects the cardiovascular, hepatic, renal and neurological systems, Ni causes decrease in

sperm count, motility and loss of fertility in males and Pb is a well known neurotoxin. Thus, Wastewater application in agricultural fields can be a way of wastewater utilization but should only be used after proper treatment to remove the contaminants for prevention of accumulation in the soil and further uptake in the food chain.

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